nature portfolio

Peer Review File

National and transboundary contributions to surface ozone concentration across European countries

Corresponding Author: Mr Roger Garatachea

Version 0:

Decision Letter:

** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors **

Dear Mr Garatachea,

Your manuscript titled "Comprehensive assessment of national versus transboundary contributions to surface ozone across European countries" has now been seen by 2 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication but would be interested in considering a revised version that fully addresses these serious concerns.

Specifically, the two reviewers find value in the main goal of the paper but have reservations about the paper. However, Reviewer #1 does not recommend publications and is very critical about the validation of the model and its accuracy. To be published in this journal, more evidence should be provided for the model's ability to derive background concentrations and a quantitative assessment of the uncertainty to convince that the model indeed goes beyond the existing studies and provides a comprehensive and accurate assessment of the local vs global ozone concentrations. Reviewer #2 asks for a better literature review.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers.

Please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

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Please do not hesitate to contact us if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Yinon Rudich, PhD

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Summary:
This paper addresses am important topic; however, it has a major shortcoming that prevents my recommending publication. My major concern is that he authors' analysis is based on a single global modeling system (CALIOPE with other components); it is well established that simulations of the ozone distribution by global models have substantial uncertainties. The authors do evaluate their model results through comparison with observations, but that comparison is limited to total ozone concentrations. All the global models in use today rely on a great many parameterizations of the multitude of chemical and physical processes that determine the ozone distribution. Those parameterizations have been highly tuned so that model results closely approach observations; thus, evaluation of model simulations by further comparison with observations is of only limited additional value. The conclusions of the present paper rely upon the accurate, separate simulation of background ozone and ozone from local and regional photochemical production. No evaluation of the model's ability to accurately achieve this accurate, separate simulation is presented; hence the results must be considered preliminary. Communications Earth & Environment aims to publish high-quality research papers that represent significant advances bringing new insight to a specialized area of the Earth, environmental or planetary sciences. Given my above concern, I do not believe that this paper fulfills that aim. I do not identify any errors in the procedures employed in the paper, and the conclusions seem to be reasonable, i.e. qualitatively consistent with other studies and expectations. It would judge it as likely suitable for publication in a more specialized journal that focuses on model development and application.

Given below are some additional discussion of my major concern identified above and specific issues that I noted in reading the paper.
1) On line 5 of page 1, the authors assert that: "Currently, there is no method to quantify the contributions of different sources to surface O3 from observations." This assertion is not accurate. For example, using only observations Derwent and Parrish (2022) separately estimate the contributions from transported baseline ozone and regional photochemical enhancements across northwestern Europe, which is part of the region focused on in the present paper. The model utilized here does provide much more detailed results, but it could benefit from a comparison with the results of that observational-based work.
2) On page 3, line 80 the authors note that six different air quality model simulations "revealed that 45% to 65% (depending on the model) of the surface O3 prevailing in Europe originates from intercontinental transport during the summer of 2010." Other studies are cited that give even higher percentages. This is wide variability, which likely does not capture the entire model uncertainty, emphasizes that the results of the present study are quite uncertain, and simply adds one more estimate to those already published.
3) Section 3.1 summarizes the evaluation of the model simulation. The results are evaluated against observations based only on the simulated total ozone concentrations. As noted earlier, model parameterizations have been carefully tuned to minimize errors in such comparisons, so there is a degree of circular reasoning in this process. Of more interest for the present study would be some evaluation of how well the model simulates the apportionment of the total ozone between "national versus transboundary contributions to surface ozone", which is the goal of the present paper as indicated in the title. Such evaluation is difficult, but there is evidence that performance of global models in this regard has large uncertainty. For example, Fiore et al. (2014) and Zhang et al. (2020) compare two of the leading global models (GFDL-AM3 or GFDL-AM4 versus GEOS-Chem). Other studies (Skipper et al., 2021; Hosseinpour et al., 2023) conclude that combining model results with observational data is needed to improve the accuracy of simulated background ozone concentrations. Unless the global modeling system used in the present paper can be demonstrated to simulate source apportionment with less uncertainty than expected from literature comparisons, the specific results presented in this paper must be considered to be preliminary, i.e., capable of informing further research, but not providing the "comprehensive understanding of the relative significance of national and transboundary pollution contributions" that the authors note "is crucial for informed policy development."

References:
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Reviewer #2 (Remarks to the Author):

Review of the manuscript “Comprehensive assessment of national versus transboundary contributions to surface ozone across European countries” by Solé et al.

General comments

The manuscript by Solé et al. describes a very thorough study in the quantification of the different contributions to Maximum Daily 8-hour Average ozone (MDA8) in the European region. The authors make use of a tagging approach in their numerical model which allows the attribution of modelled near-surface ozone to its emitted precursors. By tagging the emissions of ozone precursors (NOx and VOC) from each individual country in their model domain, mostly based on countries in the European Union, they determine the extent to which each country influences its own ozone air quality, and the extent to which ozone in each country is influenced by transboundary transport, either from neighboring countries, from countries outside of their chosen set, from international shipping, or from hemispheric transport.

Overall, I find the study to be extremely comprehensive and thorough and the manuscript to be well-presented. My major criticism of this work is that the authors do not adequately discuss their results in the context of the existing literature. While the material in their manuscript is sufficiently novel due to its application of a tagging method at such a large scope, many of the results presented are not completely new. Proper citation of existing work not only acknowledges the authors who have already published on the topics covered by this new manuscript, it also benefits the current authors by allowing their new work to be more easily discoverable through citation databases. This is an important and timely manuscript which should be well received by the European science and policy communities, and may influence the thinking on future emission control policy in Europe. I would very much like to see this manuscript published, but only after the authors have addressed this major criticism, as well as some more detailed minor comments.

Discussion of previous literature

The authors do a reasonable job of citing existing literature in their Introduction section to provide a background for their study, but unfortunately their Results section is almost completely free of any discussion of how the new results compare to the papers which are cited in the Introduction. Two key examples here are Lupaşcu and Butler (2019) and Romero-Alvarez et al. (2022). Both of these studies are correctly discussed in the Introduction, but not mentioned at all in the discussion of the results. Both of these studies overlap significantly with the new work, in that they use tagging methods to quantify national and transboundary contributions to ozone in the European region. Neither of these studies has such a large scope as the new work, but the degree of overlap with the new work warrants some additional discussion. The work of Romero-Alvarez et al. (2022) focuses on the United Kingdom, so could be discussed in the parts of the Results section that focus on the contributions to ozone there. The work of Lupaşcu and Butler (2019) actually covers a similar domain to the new work, but is less ambitious in scope due to its use of aggregated regions instead of individual countries, and its neglect of interannual variability. Nevertheless, the new results should be discussed in the context of this previous work. For example, many of the regions in Lupaşcu and Butler (2019) correspond broadly with the regions used in the new work, and the relative contributions of precursor emissions from the “local” region, neighboring countries, international shipping, and hemispheric transport. Furthermore, the earlier work of Lupaşcu and Butler (2019) also discusses how these contributions change depending on the metric. Crucially, they also show that for the 95th percentiles, the local and nearby contributions are larger than in the average case. There are numerous places in the Results section of this new work that touch on these points, but no discussion of the relevant previous work is given.

Another interesting result of the authors’ new work is the large contribution of hemispheric transport to European ozone

levels. This is also not new. The authors do indeed cite several relevant papers in their Introduction for this (HTAP, 2010; Jonson et al., 2018; Lupaşcu and Butler, 2019; Romero-Alvarez et al., 2022; Zohdirad et al., 2022), but in the results section of the new work where this long-range contribution is quantified, this result is barely discussed in the context of the previous work. Another highly relevant publication the authors should be aware of on this topic is Butler et al. (2020), which while global in scope does have things to say about the European region and also makes use of a tagging methodology.

Yet another under-discussed result is the influence of international shipping on European ozone. This is not mentioned in the Introduction section at all, and only one previous study is cited on this (Petetin et al., 2023), which is only cited in the conclusions section, with no discussion of why this study is significant. At the very least, there should be a paragraph in the Introduction on this subject, and the Results section should mention the previous literature where appropriate and discuss the new results in that context. There are also several other relevant studies here (Eyring et al., 2007; Karl et al., 2019; Lupaşcu and Butler, 2019; Butler et al., 2020; Jonson et al., 2020; Schwarzkopf et al., 2022; Zohdirad et al., 2022) as a non-exhaustive list.

Detailed comments

Section 2.1: Are the ozone concentrations reported at the lowest model level, or are they extended to the surface? For maximum policy-relevance, ozone values at 2m should be reported. More information on the model vertical structure is needed here, including the height of the lowest model level.

Lines 148-151: It would be useful to give some examples of the different kinds of tagging systems listed here. Also, there is one more option that is not listed here: simultaneous full attribution to both NOx and VOC reactants. This is the approach taken by Lupaşcu and Butler (2019), and it avoids the disadvantages of the pure forms of options 2 or 3, at the cost of increased difficulty of interpreting the results.

Line 155: What are the implications of the limiting-precursor approach as chosen here in comparison to the other possible tagging methods?
• The method would be sensitive to the chemical regime as simulated by the model. How well does the model reproduce NOx-limited or VOC-limited regimes? How does it compare to other models in simulating the chemical regime?
• In VOC-limited conditions, some fraction of the ozone production will be attributed to biogenic VOCs and methane (which has a longer lifetime but is ~1000 times more concentrated than other VOCs).

Line 164: Why does this method lead to a higher hemispheric contribution? A short explanation would be useful here.

Line 165: How is methane handled in the ozone attribution? Is it contained in one of the species listed in Table S1? The CCAC Global Methane Assessment (CCAC, 2021) shows that ozone production is sensitive to methane in regions with high NOx, which includes much of Europe. How is ozone production from methane attributed in the current study?

Line 168: Does Figure 1 show the actual model domain? Latitude and Longitude scales should be added.

Line 172: Part of the ozone attributed to each country in this study will be from BVOC emissions, which may be beyond the control of policymakers. One might argue that this will only happen under VOC-limited conditions (where the excess NOx is from anthropogenic sources), but in this case, wouldn't it be better to attribute that ozone to the anthropogenic NOx emissions for the purposes of policy advice?

Section 3.1: There is no discussion of how well the boundary condition from the global model represents the inflow of ozone into the model domain. Given that hemispheric transport is such a major contributor to ozone in this study, some discussion of this is needed, especially since there is a large inter-model spread in near-surface ozone from current state-of-the-art global models (Griffiths et al., 2021). Perhaps the authors could highlight the evaluation of their simulated ozone at the Mace Head station, which is primarily influenced by long-range transport.

Line 244: Please check the grammar in this sentence, it doesn't seem to make sense to me.

Lines 390-400: This would be a good place in the manuscript to add some discussion of how these new results compare with previous work on the influence of international shipping on European ozone.

Line 413: The Gothenburg Protocol is not global. Its geographical scope is limited to the UNECE region.

Lines 417-418: It is not clear from this text or from the caption to Figure 7 what exactly is shown here. Please explain this more clearly.

Line 447: The "potential to address" high ozone levels is problematic here. The authors seem to mean that if the contribution is high, then there must be a high potential for reduction, but this is not how policymaking works. Other factors are also important, such as whether the current emissions are due to the best available technology or not, what the costs of mitigation would be relative to the benefits of meeting regulatory targets, and whether or not any reductions would actually contribute to meeting these targets. I don't expect the authors to go into this level of detail, but I think they should define what they mean by "potential" here.

Line 473: Part of the reason why Western European countries tend to have a higher contribution to their own ozone levels

could be due to the simple fact that they often tend to be larger, so that ozone production downwind of emission sources is more likely to take place within the national boundaries. This is not really discussed here.

Figure 9: The transition from ozone-exporting to ozone-importing countries appears to generally follow a west-east gradient. Countries in Western Europe still do import ozone, but because of their geographical location, this ozone is tagged as “BCON”. The prevailing westerly winds then lead to ozone export from Western Europe to Eastern Europe. This pattern can also be seen in Figure 2. The way that the authors present this ozone import/export is still useful as a kind of “blame matrix”, but the role of geography on this result should be better discussed.

Line 568: I do not understand this sentence. Please clarify this.

References

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Author Rebuttal letter: The author's response to these comments can be found at the end of this file.

Version 1:

Decision Letter:

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Dear Mr Garatachea,

Your manuscript titled "Comprehensive assessment of national versus transboundary contributions to surface ozone across European countries" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Best regards,

Yinon Rudich, PhD
Editorial Board Member
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Martina Grecequet, PhD
Associate Editor,
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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have put great effort into addressing my concerns, including:
• Additional comparisons between the model results and observations at both remote and regional background EMEP stations across Europe, and at the stations with the largest European anthropogenic contribution.
• A comparison of the model results with the analysis of Derwent and Parrish (2022).
The latter comparison is quite persuasive to me, given that the results are derived from such different approaches. However, I must argue a bit with one paragraph in their rebuttal (which is also included in the revised Supplement):

“We note that while Derwent and Parrish (2022) computed RPE for the entire year 2020, our study focuses exclusively on the summer months of 2015, 2016, and 2017. Therefore, consistent with the heightened RPE during summer conditions due to higher temperatures and solar radiation, we obtain higher RPE values compared to those estimated by Derwent and Parrish (2022), whose RPE spans the entire year. However, despite the disparity in analysis periods (annual vs summer), we find a strong correlation across the 32 stations between our simulated RPE and the observationally-derived values (R2 = 0.80), as depicted in Fig. 3.”

Derwent and Parrish do consider observational data from the entire year, but the RPE they calculate is based upon annual maximum 8-hourly (AM8) mean ozone mixing ratios. These annual maxima predominately occur in summer, so the differences that are seen in the comparison cannot really be attributed to the disparity in analysis periods (annual vs summer). I suggest that this paragraph in the Supplement be revised accordingly.

These added comparisons, the comments of the second reviewer, combined with the authors’ rather extensive revisions compel me to reverse my previous opinion, and to now support publication of this paper.

Reviewer #2 (Remarks to the Author):

The authors have done a good job of responding to the comments of both reviewers, and of revising their manuscript accordingly. The resulting manuscript is of high quality. I somewhat agree with the criticism of Reviewer 1, that the submission may not fully satisfy the stated scope of Communications Earth & Environment, that “Research papers published by the journal represent important advances bringing new insight to a specialized area of research.” (quote from the Guide to authors on the journal website). None of the individual insights from the work represent especially important advances. Nevertheless, I also see value in the authors’ response that the sheer breadth of their new work goes well beyond any of the individual studies that have been published on this topic. I would definitely like to see this work published, but I am not completely sure whether Communications Earth & Environment is the most appropriate journal for this. I would leave the final decision on this to the professional editorial team of the journal.

Author Rebuttal letter: The author's response to these comments can be found at the end of this file.

Response to Reviewer 1

We thank the reviewer for the insightful comments, which have helped us complementing and improving the manuscript. A detailed point-by-point response to all the questions, comments, and suggestions raised by the reviewer is provided below.

E.1. Summary: This paper addresses an important topic; however, it has a major shortcoming that prevents my recommending publication. My major concern is that the authors' analysis is based on a single global modeling system (CALIOPE with other components); it is well established that simulations of the ozone distribution by global models have substantial uncertainties.

Thank you for recognizing the relevance of our work. We acknowledge the reviewer's concern, which may stem partly from a misunderstanding of the characteristics of the modeling system employed in our study. To provide clarity, we describe below how our study draws upon multiple modeling components strategically integrated to minimize uncertainties that may arise from relying solely on a global model:

1) **High-resolution regional modeling**: Our study relies on a regional chemical transport model, not a global model. The CALIOPE system is specifically designed to accurately describe the lifecycle of key atmospheric chemistry species, such as O_3, in the troposphere over Europe. In contrast to global models, which often face challenges due to coarse grid resolutions and simplified chemical mechanisms, CALIOPE focuses on the European region with a detailed and state-of-the-art chemical mechanism and high resolution. Its primary objective is to refine global estimates, providing significantly more reliable information for air quality studies within the European context. We would like to emphasize that CALIOPE regional simulations have undergone rigorous evaluation in multiple studies at high spatial and temporal scales (Baldasano et al., 2008; Pay et al., 2010, 2014, 2019), consistently demonstrating its effectiveness in capturing atmospheric dynamics and pollutant behavior. As outlined in Section 2.1, our study employed the CALIOPE system over a European domain at a resolution of 18 x 18 km^2, enabling detailed assessments of tropospheric O_3 concentrations at surface level. Additionally, in contrast to the less precise emission inventories typically used in global models, we utilized a recently developed anthropogenic emission inventory specifically tailored to the European region (Kuenen et al., 2022), which enhances the fidelity of our modeling framework to real-world conditions.

2) **Observationally constrained boundary conditions**: Our regional modeling system benefits from observationally constrained boundary conditions for both meteorology and chemistry. Specifically, we integrate meteorological reanalysis data from the European Center for Medium-range Weather Forecast (ECMWF) to establish boundary conditions for meteorological parameters. Additionally, for chemical boundary conditions, we utilize the CAMS global analysis provided by the Copernicus programme. This dataset is notable for its assimilation of data from various satellite sensors, including OMI, SBUV, GOME-2, MLS, OMPS for O_3 assimilation, IASI and MOPITT for CO, and OMI for NO2. Consequently, our approach ensures that the contribution of hemispheric sources to ozone levels in Europe is substantially constrained by observational data, thereby enhancing the reliability and accuracy of our modeling outcomes, compared to unconstrained global modeling studies.

We have further clarified these aspects in the Methodology section when introducing the CALIOPE system as follows:

L123: "We used the CALIOPE regional air quality model system to simulate the surface O_3 concentrations over Europe."

L134-137: "We note that the CAMS analysis is notable for its assimilation of data from various satellite sensors, including OMI, SBUV, GOME-2, MLS, OMPS for O_3 assimilation, IASI and MOPITT for CO, and OMI for NO_2. Consequently, our approach ensures that the contribution of hemispheric sources to ozone levels in Europe is substantially constrained by observational data, thereby enhancing the reliability and accuracy of our modeling outcomes."

E.2. The authors do evaluate their model results through comparison with observations, but that comparison is limited to total O_3 concentrations. All the global models in use today rely on a great many parameterizations of the multitude of chemical and physical processes that determine the O_3 distribution. Those parameterizations have been highly tuned so that model results closely approach observations; thus, evaluation of model simulations by further comparison with observations is of only limited additional value.

We acknowledge the reviewer's concern regarding the limitations of the parametrizations used to simulate the chemical and physical processes in models. While models rely on ongoing refinement and tuning of these parametrizations to mitigate inherent uncertainties, comparisons with independent observations are a fundamental step for

both advancing our understanding and provide confidence on model results. Our model incorporates state-of-the-art parameterizations of atmospheric chemistry, which represents to a large extent the current understanding of these processes.

It is relevant to emphasize that O_3 plays a pivotal role as a key reactive species of secondary origin, exerting significant control over the oxidative capacity of the atmosphere. Rather than being directly emitted, O_3 is formed through intricate photochemical reactions, impacting a plethora of reactive chemistry processes within the troposphere. Assessing O_3 in models with observations offers a robust overall evaluation of the representation of various complex processes incorporated into the model, such as emissions, transport, chemical transformation, and sinks. In our work, the model results were rigorously tested by two different evaluation methods against observational data proposed by the scientific community (Emery et al., 2017; Janssen and Thunis, 2022), showing a good agreement between modelled and observed O_3 concentration.

We firmly believe that our model evaluation is thorough and instills confidence in the results presented in the study. Yet, to further address the reviewer's concern, we have significantly expanded our evaluation in the revised version of the manuscript. We have incorporated additional analyses to discriminate and evaluate modeled background O_3 concentrations and O_3 from photochemical regional production against observation-based estimates, which provides additional evidence of the robustness of our study. A detailed account of the complementary evaluation is provided below in response to the reviewer´s comments E3 and E5.

E.3. The conclusions of the present paper rely upon the accurate, separate simulation of background ozone and ozone from local and regional photochemical production. No evaluation of the model's ability to accurately achieve this accurate, separate simulation is presented; hence the results must be considered preliminary.

We thank the reviewer for this comment. As suggested by the reviewer in comment E5 below, some observation-based studies quantify the contribution of background ozone and regional photochemical production by analysing O_3 concentrations measured in stations representative of background conditions in contrast to stations also affected by local/regional photochemistry. This serves as an approximation to infer both the background and the photochemical enhancement over specific regions based on measurements. Although this method is not

devoid of uncertainties, it provides an estimate of the background O_3 contribution to total O_3. Therefore, to address the reviewer's concern, we have expanded the evaluation of the model in the revised manuscript, introducing additional analyses disentangling background O_3 concentrations and photochemical enhancements.

For such evaluation, we used O_3 concentrations obtained from remote and regional background EMEP stations across Europe. We specifically assessed the model's performance at stations where the simulated hemispheric contribution is at its peak, as well as stations where the model exhibits the greatest national influence (Fig. 1). This approach allowed scrutinizing the model's ability to capture background conditions and local/regional photochemical enhancements, respectively.

As depicted in Fig. 1, stations mainly influenced by background O_3 (Max BCON) are located along the Atlantic coast. On average there is a positive bias which remains below 17 % (Fig. 2). For reference, our model closely aligns with the results obtained by the Copernicus Programme CAMS global reanalysis (CAMSRA), which is one of the best products available to describe O_3 concentrations at a global scale as it combines both model and observation-based information to provide a comprehensive estimation of the atmospheric conditions (Inness et al., 2019). On the other hand, the model presents a very good agreement in stations heavily influenced by local/regional photochemical production (Max National) (Fig. 2). Most notably, this comparison highlights that overall the model captures well the transition from background conditions to photochemical O_3 production regimes, indicating its ability to reproduce O_3 variability effectively.
The good agreement between the background and photochemical increment derived from observations and the numerical simulations instills again stronger confidence in all the discussion and findings of our results. We thank the reviewer for the suggestion as this additional level of evaluation further strengthens the findings of our study and supports the methods employed.

Note that in response to another related specific comment from the reviewer below, we have additionally performed a direct comparison with the observation-based estimates from Derwent and Parrish (2022), who separately estimated the contributions from transported baseline ozone and regional photochemical enhancements across northwestern Europe (see below).

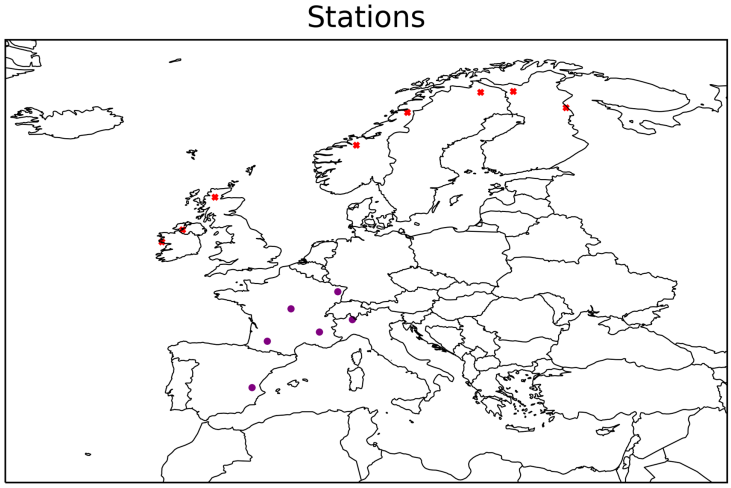

Figure 1: Geographical distribution of the stations with contributions from the hemispheric O_3 (BCON) above the 90^{th} percentile (red) and stations with national contributions to O_3 above the 90^{th} percentile (purple).

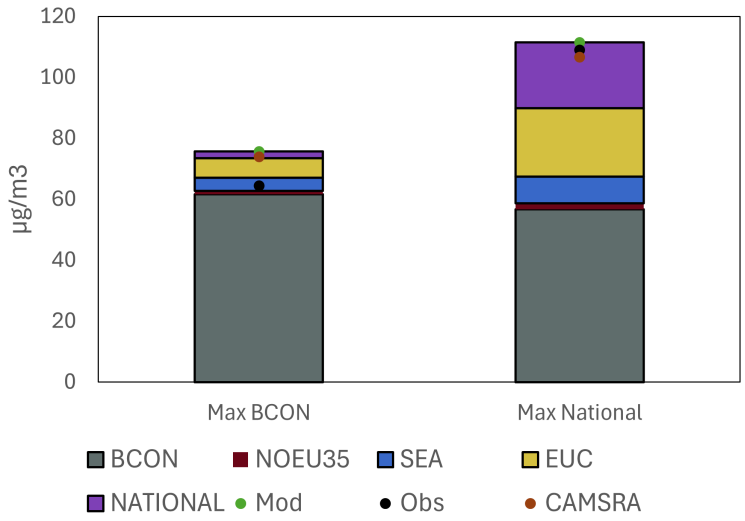

Figure 2: Average simulated source contributions to O_3 and total simulated and measured O_3 in EMEP stations where the contributions of hemispheric O_3 are above the 90^{th} percentile (Max BCON) and where the national contributions are above the 90^{th} percentile (Max National) of all EMEP stations. The geographical distribution of those stations is presented in Figure 1. Contributions include the national O_3 contribution (NATIONAL), the imported O_3 in each country originated from each of the remaining 34 countries (EUC), the contributions from hemispheric transport (BCON), non-European neighboring countries (NOEU35) and maritime sources (SEA). The green dot represents the model total O_3 concentration, the black dot the O_3 concentrations from observations, and the red dot the O_3 concentrations from the CAMS reanalysis (CAMSRA).

The results of this extended evaluation are now included in the supplementary material (new Sect. 5 in the supplement) and discussed in the main text.

In the *Observations and model evaluation* section, we describe the methodological aspects of the background and photochemical enhancements as follows:

L226-232: "Additionally, we specifically assessed the model's performance at stations where the simulated hemispheric contribution is at its peak, as well as at stations where the model exhibits the greatest national influence (Sect. S5). For this assessment we used the remote and regional background European Monitoring and Evaluation Programme (EMEP) stations (Tørseth et al., 2012) within the AQ (Air Quality) eReporting database of the EEA. This approach allows scrutinizing the model's ability to capture background conditions and local/regional photochemical enhancements, respectively. This comparison also uses as a reference the Copernicus Programme CAMS global reanalysis (CAMSRA), which combines both model and observation-based information to provide a comprehensive estimation of the atmospheric conditions (Inness et al., 2019; Lacima et al., 2023)."

Furthermore, a discussion is now included in the *Evaluation of simulated O_3 concentrations* section as follows:

L263-265: "Additionally, the model captures well the increase in MDA8 O_3 concentrations between EMEP stations where the contributions from hemispheric transport O_3 is most dominant and those that are strongly impacted by national contributions, reflecting the ability of the model to represent local to regional photochemical O_3 enhancements (Sections S.5 and S.6 in the supplement).

E.4. Communications Earth & Environment aims to publish high-quality research papers that represent significant advances bringing new insight to a specialized area of the Earth, environmental or planetary sciences. Given my above concern, I do not believe that this paper fulfills that aim. I do not identify any errors in the procedures employed in the paper, and the conclusions seem to be reasonable, i.e. qualitatively consistent with other studies and expectations. It would judge it as likely suitable for publication in a more specialized journal that focuses on model development and application.

We believe that our paper offers valuable insights that align well with the aims of *Communication Earth & Environment*. On the one side, above and in the specific comments that follow we have thoroughly addressed the key reviewer's concerns by adding specific evaluation that reinforces the robustness of our approach. On the other side, we would like to highlight the added value compared with other studies. Our paper stands out in the literature due to its comprehensive spatial and temporal coverage and detailed analysis of the transboundary O_3 contributions across 35 countries in Europe, encompassing aspects such as import-export dynamics, interannual variability, and the contributions of strong episodes, many of which have been barely explored in such depth in previous studies. We believe these contributions, combined with the improved evaluation of the model results, make our paper particularly compelling for the readers of *Communications Earth & Environment*.

E.5. On line 5 of page 1, the authors assert that: "Currently, there is no method to quantify the contributions of different sources to surface O_3 from observations." This assertion is not accurate. For example, using only observations Derwent and Parrish (2022) separately estimate the contributions from transported baseline ozone and regional photochemical enhancements across northwestern Europe, which is part of the region focused on in the present paper. The model utilized here does provide much more detailed results, but it could benefit from a comparison with the results of that observational-based work.

Thanks for this comment. Our assertion referred to the fact that there is no "direct" observational method to disentangle the contributions from different sources. However, we agree with the reviewer that there are observation-based analyses that allow estimating contributions of transported baseline ozone and regional photochemical enhancements and that our statement could be confusing. Note that we also referred to the multiple sources that contribute to O_3 formation, not only the transported hemispheric contribution and the regional photochemical enhancements. We have removed the sentence from the abstract.

We also recognize the relevance of works such as Derwent and Parrish (2022), which estimated the long-range transport of O_3 (using the station of Mace Head as a reference) and the regional photochemical contribution over northwestern Europe using an observation-based analysis and non-linear regression techniques. Derwent and Parrish (2022) showed that the hemispheric contribution controls O_3 in northwestern Europe, estimating that around 85% of the annual average of the maximum daily 8-hour running mean concentration is due to long-range transport in the United Kingdom. This result aligns well with our findings and previous modeling works by Romero-Alvarez et al. (2022); Zohdirad et al. (2022), with the three studies finding that long-range transport contributes around 70-90% in the United Kingdom during the summer months.

Derwent and Parrish (2022) computed regional photochemical enhancement (RPE) values for year 2020 at each of the low-elevation EMEP stations in northwestern Europe (32 stations). The RPE quantifies the photochemical production driven by local and regional anthropogenic precursor emissions. Following the reviewer´s suggestion, we conducted a comparative analysis of RPE values at these EMEP stations, derived from our simulations, with those reported by Derwent and Parrish (2022) (see Table 1). In our model-derived RPE estimation we incorporate contributions from national sources, imported O_3 originating from the remaining 34 countries (EUC), and

maritime sources (SEA). We note that while Derwent and Parrish (2022) computed RPE for the entire year 2020, our study focuses exclusively on the summer months of 2015, 2016, and 2017. Therefore, consistent with the heightened RPE during summer conditions due to higher temperatures and solar radiation, we obtain higher RPE values compared to those estimated by Derwent and Parrish (2022), whose RPE spans the entire year. However, despite the disparity in analysis periods (annual vs summer), we find a strong correlation across the 32 stations between our simulated RPE and the observationally-derived values ($R^2 = 0.80$), as depicted in Fig. 3. Such comparison with observational data further substantiates the robustness of our modeling method to disentangle contributions from long-range transport and photochemical production driven by local and regional anthropogenic precursor emissions.

Complementing the new material introduced in the evaluation section of the revised manuscript, we added the RPE discussion in the revised manuscript too, and in the supplementary material (Sect. S6). In the *Observations and model evaluation* section, we describe the methodological aspects of RPE calculations as follows:

L234-236: "Finally, we compared our results against the observation-based analysis of Derwent and Parrish (2022), where the contributions of long-range transport and regional photochemical enhancements (RPE) to O_3 were estimated for EMEP stations in northwestern Europe (see Sect. S6 for details)."

A comment is included in the *Evaluation of simulated ozone concentrations* section as follows:

L266-269: "We also found a strong correlation between the observation-based estimates from Derwent and Parrish (2022) and our simulated values across 32 EMEP stations in northwestern Europe ($R^2 = 0.80$, see Sect. S6). Such comparison with observational data further substantiates the robustness of our modeling method to disentangle contributions from long-range transport and photochemical production driven by local and regional anthropogenic precursor emissions."

Table 1: Comparison of Regional Photochemical Enhancement (RPE) values from Derwent and Parrish (2022) in 2020 (annual) and CALIOPE in 2015, 2016 and 2017 (summer). The RPE in CALIOPE is approximated as the contributions from national sources, imported O_3 originating from the remaining 34 countries (EUC), and maritime sources (SEA)

Station code	**Longitude,** °E	**Latitude,** °N	**RPE 2020** Derwent and Parrish (2022) (ppb)	**RPE CALIOPE** (ppb)
AT0002R	16.767	47.767	21	33.48
AT0042R	15.047	48.879	21	31.57
AT0043R	15.919	48.106	26	35.22
AT0045R	15.547	48.371	20	33.55
AT0046R	16.731	48.335	22	32.51
AT0047R	16.677	48.051	24	33.22
BE0032R	6.003	50.629	34	30.42
CH0002R	6.945	46.813	22	26.28
CH0003R	8.905	47.48	26	31.05
DE0001R	8.31	54.926	18	25.69
DE0002R	10.759	52.802	26	30.78
DE0007R	13.033	53.167	18	28.61
DE0009R	12.733	54.433	18	27.20
DK0031R	8.433	56.283	16	19.85
DK0041R	12.126	55.687	6	20.82
FI0009R	21.377	59.779	9	21.41
FI0017R	27.686	60.527	12	16.91
FI0022R	29.402	66.32	5	7.02
IE0031R	9.5	53.167	5	6.09
IT0004R	8.633	45.8	28	38.17
NL0009R	6.277	53.334	12	24.65
NL0010R	5.854	51.451	25	30.52
NO0001R	8.25	58.383	8	13.91
NO0015R	13.917	65.833	9	8.04
NO0039R	8.883	62.783	8	9.74
NO0042G	11.883	78.9	0	1.65
NO0043R	11.533	59	10	14.92
SE0011R	13.15	56.017	13	20.67
SE0012R	17.383	58.8	10	16.43
SE0013R	21.067	67.883	6	6.37
SE0032R	15.567	57.817	13	15.80
SE0035R	19.767	64.25	6	9.03

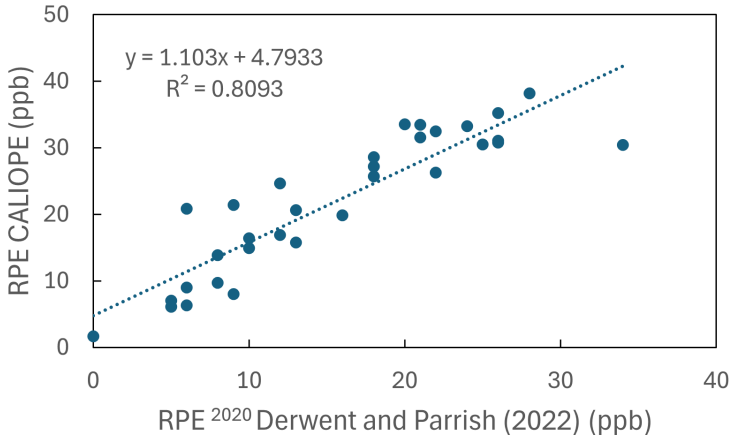

Figure 3: Linear relationship and coefficient of determination (R^2) between the RPE found by Derwent and Parrish (2022) and by the CALIOPE system across 32 EMEP stations in northwestern Europe.

E.6. On page 3, line 80 the authors note that six different air quality model simulations "revealed that 45% to 65% (depending on the model) of the surface O_3 prevailing in Europe originates from intercontinental transport during the summer of 2010." Other studies are cited that give even higher percentages. This is wide variability, which likely does not capture the entire model uncertainty, emphasizes that the results of the present study are quite uncertain, and simply adds one more estimate to those already published.

The reviewer's acknowledgment of differences across studies underscores the intricate nature and inherent uncertainties in modeling. In response, we've meticulously revised our manuscript, augmenting the literature review to underscore the importance of mitigating these uncertainties while also highlighting the consistency between our findings and prior research (Jonson et al., 2018; Zohdirad et al., 2022; Derwent and Parrish, 2022; Romero-Alvarez et al., 2022). It's noteworthy that this consistency, despite methodological disparities across studies, serves as a validation of our work.

In our revised manuscript, we've enriched the discussion with additional contextualization, providing readers with a deeper understanding of our results vis-à-vis the existing literature. This not only shows the robustness of our findings but also enhances the utility of our study. Moreover, we must note that part of the differences in literature may stem from natural interannual variability, as different studies focus on distinct annual periods. To this end, our study offers a novel perspective by comprehensively examining the role of this variability.

Furthermore, our research marks a significant advancement in the field. We provide estimations previously unavailable, such as a detailed analysis spanning 35 European countries and an exploration of import-export dynamics between nations. This expansion of scope not only enriches the discourse but also lays a foundation for future investigations in understanding the complexities of source apportionment of O_3.

This sentence, which was already in the *Introduction* section, highlights the variability of O_3 from intercontinental transport:

L72-75: "Current estimates of the external contribution to O_3 in Europe show a notable range of variability, likely arising from diverse chemical transport models (CTMs), methodologies for source apportionment, input data sets, and interannual variability, among other factors, used across studies."

In the revised manuscript, we have introduced further

discussion with previous literature findings as follows:

L70-72: "Additionally, through an observation-based analysis, Derwent and Parrish (2022) estimated that approximately 85% of the annual 8-hourly maximum ozone levels in 2020 in the United Kingdom were attributable to long-range transport."

L278-280: "BCON contributes strongly to O_3 in all European countries, ranging from 37% in Malta to 88% in Iceland of the total MDA8 O_3, consistent with prior studies (Jonson et al., 2018; Zohdirad et al., 2022). "

L291-292: "Indeed, Romero-Alvarez et al. (2022) reported similar contributions in the United Kingdom, where the majority of O_3 comes from hemispheric transport during May-August 2015."

L311-322: "Previous studies (HTAP, 2010; Jonson et al., 2018; Lupaşcu and Butler, 2019; Butler et al., 2020; Romero-Alvarez et al., 2022; Zohdirad et al., 2022) consistently underscore the significant role of long-range transport on O_3 levels in Europe, aligning with our study. However, the percentage attributed to long-range transport of O_3 varies across studies, where most of them range from 45% to 65%. Notably, Lupaşcu and Butler (2019) generally report lower attribution to long-range transport and higher attribution to the local and rest of Europe's contribution compared to our findings. This discrepancy may be attributed to differences in the tagging methodology employed by Lupaşcu and Butler (2019), where O_3 formation is exclusively attributed to NOx emissions (see Section 2.2). Shu et al. (2022) reports a similar trend when they compared different tagging methodologies, with a significant reduction of long-range transport contribution when only NOx emissions are tagged. However, Romero-Alvarez et al. (2022), with the same approach as Lupaşcu and Butler (2019), in a more regional study in the United Kingdom, found results more aligned with our findings. Derwent and Parrish (2022) also showed results consistent with our findings, showing alignment between results derived from an observational-based analysis and modeled results (See Sect. S6 in the Supplement)."

E.7. Section 3.1 summarizes the evaluation of the model simulation. The results are evaluated against observations based only on the simulated total ozone concentrations. As noted earlier, model parameterizations have been carefully tuned to minimize errors

in such comparisons, so there is a degree of circular reasoning in this process. Of more interest for the present study would be some evaluation of how well the model simulates the apportionment of the total ozone between "national versus transboundary contributions to surface ozone", which is the goal of the present paper as indicated in the title. Such evaluation is difficult, but there is evidence that the performance of global models in this regard has large uncertainty. For example, Fiore et al. (2014) and Zhang et al. (2020) compare two of the leading global models (GFDL-AM3 or GFDL-AM4 versus GEOS-Chem). Other studies (Skipper et al., 2021; Hosseinpour et al., 2023) conclude that combining model results with observational data is needed to improve the accuracy of simulated background ozone concentrations. Unless the global modeling system used in the present paper can be demonstrated to simulate source apportionment with less uncertainty than expected from literature comparisons, the specific results presented in this paper must be considered to be preliminary, i.e., capable of informing further research, but not providing the "comprehensive understanding of the relative significance of national and transboundary pollution contributions" that the authors note "is crucial for informed policy development."

Regarding the use of global models in our study, we have clarified this misunderstanding in response to comment E.1. In our work, we use a regional modeling system constrained by both meteorological and chemical analyses in the boundaries. Please see the discussion in comment E.1.

The limitations of the evaluation have also been addressed in responses to comments in E3 and E5 above. Following the reviewer's advice, we have complemented the evaluation presented in the original manuscript, mostly relying on two different methods accepted by the scientific community to evaluate total O_3 concentrations (Emery et al., 2017; Janssen and Thunis, 2022), with additional assessment of the skills of the model to simulate background concentrations and the photochemistry enhancement.

The consistency found in the complementary evaluation requested by the reviewer and introduced in the revised manuscript points out the robust representation of O_3 and its contributions in our simulation over Europe. We are confident that our work represents a significant step forward with new estimates and discussions not provided before in the literature, such as the detailed analysis across 35 European countries, the interannual variability of the different source contributions, and the import-export dynamics between countries.

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Response to Reviewer 2

We thank the reviewer for the constructive review and we appreciate the reviewer's recognition of our paper's value to the scientific and policy communities. A detailed point-by-point response to all the questions, comments, and suggestions raised by the reviewer is provided below.

General comments:

E.1. My major criticism of this work is that the authors do not adequately discuss their results in the context of the existing literature. While the material in their manuscript is sufficiently novel due to its application of a tagging method at such a large scope, many of the results presented are not completely new. Proper citation of existing work not only acknowledges the authors who have already published on the topics covered by this new manuscript, it also benefits the current authors by allowing their new work to be more easily discoverable through citation databases. This is an important and timely manuscript which should be well received by the European science and policy communities and may influence the thinking on future emission control policy in Europe. I would very much like to see this manuscript published, but only after the authors have addressed this major criticism, as well as some more detailed minor comments.

We sincerely appreciate the reviewer for recognizing the significance of the research topic and the importance of our study. We acknowledge the need of thoroughly discussing our results in relation to the existing literature, a point highlighted by the reviewer. While our original manuscript offered a comprehensive literature review that motivated the objective of our work in the introduction, we acknowledge the need for a deeper discussion of our results in relation to prior studies.

In response to this valuable feedback, we have extensively addressed this concern in the revised manuscript. Specifically, in response to comments E2, E3, and E4, we provide detailed descriptions of how we have enriched the discussion by incorporating our findings within the broader context of existing literature. We believe that these revisions significantly enhance the depth and comprehensiveness of our manuscript, providing readers with a more nuanced understanding of our research findings.

Discussion of previous literature

E.2. The authors do a reasonable job of citing existing literature in their Introduction section to provide a background for their study, but unfortunately their Results section is almost completely free of any discussion of how the new results compare to the papers which are cited in the Introduction. Two key examples here are Lupaşcu and Butler (2019) and Romero-Alvarez et al. (2022). Both of these studies are correctly discussed in the Introduction, but not mentioned at all in the discussion of the results. Both of these studies overlap significantly with the new work, in that they use tagging methods to quantify national and transboundary contributions to ozone in the European region. Neither of these studies has such a large scope as the new work, but the degree of overlap with the new work warrants some additional discussion. The work of Romero-Alvarez et al. (2022) focuses on the United Kingdom, so could be discussed in the parts of the Results section that focus on the contributions to ozone there. The work of Lupaşcu and Butler (2019) actually covers a similar domain to the new work but is less ambitious in scope due to its use of aggregated regions instead of individual countries, and its neglect of interannual variability. Nevertheless, the new results should be discussed in the context of this previous work. For example, many of the regions in Lupaşcu and Butler (2019) correspond broadly with the regions used in the new work, and the relative contributions of precursor emissions from the "local" region, neighboring countries, international shipping, and hemispheric transport. Furthermore, the earlier work of Lupaşcu and Butler (2019) also discusses how these contributions change depending on the metric. Crucially, they also show that for the 95^{th} percentile, the local and nearby contributions are larger than in the average case. There are numerous places in the Results section of this new work that touch on these points, but no discussion of the relevant previous work is given.

Following the reviewer's comment, we have revised both the *Introduction* and *Results* section to include relevant literature citations to enrich, on the one hand, the motivation of our work and, on the other hand, to put our results in the perspective of previous findings in the literature.

In the *Introduction* section, we have complemented the discussion on the hemispheric contribution or background O_3 concentrations to surface O_3 levels in Europe, introducing an observation-based estimate from Derwent and Parrish (2022):

L70-72: "Additionally, through an observation-based analysis, Derwent and Parrish (2022) estimated that approximately 85% of the annual 8-hourly maximum ozone levels in 2020 in the United Kingdom were attributable to long-range transport."

Additionally, we have also introduced a paragraph in the *Introduction* discussing the relevance of maritime emissions as an important source of O_3 (see response E4 for further details).

Regarding the *Results* section and following the suggestions of the reviewer, the revised manuscript now includes further discussion of our results compared with previous scientific findings related to the national contribution, the increase of the national contribution in high O_3 episodes, and the long-range transport.

L278-280: "A key result shown in Fig. 2 is the dominant contribution from long-range transport (BCON) to all receptor regions. BCON contributes strongly to O_3 in all European countries, ranging from 37% in Malta to 88% in Iceland of the total MDA8 O_3, consistent with prior studies (Jonson et al., 2018; Zohdirad et al., 2022)."

L291: "Indeed, Romero-Alvarez et al. (2022) reported similar contributions in the United Kingdom, where the majority of O_3 comes from hemispheric transport during May-August 2015."

L343: "This aligns with the findings of Lupaşcu and Butler (2019), who reported an increase in the NATIONAL contribution for O_3 above the 95^{th} percentile."

L365: "Notably, Pay et al. (2019) also found an increase of NATIONAL O_3 during high O_3 episodes in Spain."

L478: "These results are aligned with the findings of Romero-Alvarez et al. (2022) during May-August 2015 in the United Kingdom, who reported an average NATIONAL contribution of 13%."

E.3. Another interesting result of the authors' new work is the large contribution of hemispheric transport to European ozone levels. This is also not new. The authors do indeed cite several relevant papers in their Introduction for this (HTAP, 2010; Jonson et al., 2018; Lupaşcu and Butler, 2019; Romero-Alvarez et al., 2022; Zohdirad et al., 2022), but in the results section of the new work where this long-range contribution is quantified, this result is barely discussed in the context of the previous work. Another highly relevant publication the authors should be aware of on this topic is Butler et al. (2020), which while global in scope does have things to say about the European region and also makes use of a tagging methodology.

We acknowledge the importance of hemispheric transport to European O_3 concentrations and the uncertainties associated with its quantification. Following the reviewer suggestion, we have added a new paragraph in the *Results* section to address this point:

L311-322: "Previous studies (HTAP, 2010; Jonson et al., 2018; Lupaşcu and Butler, 2019; Butler et al., 2020; Romero-Alvarez et al., 2022; Zohdirad et al., 2022) consistently underscore the significant role of long-range transport on O_3 levels in Europe, aligning with our study. However, the percentage attributed to long-range transport of O_3 varies across studies, where most of them range from 45% to 65%. Notably, Lupaşcu and Butler (2019) generally report lower attribution to long-range transport and higher attribution to the local and rest of Europe's contribution compared to our findings. This discrepancy may be attributed to differences in the tagging methodology employed by Lupaşcu and Butler (2019), where O_3 formation is exclusively attributed to NOx emissions (see Section 2.2). Shu et al. (2022) reports a similar trend when they compared different tagging methodologies, with a significant reduction of long-range transport contribution when only NOx emissions are tagged. However, Romero-Alvarez et al. (2022), with the same approach as Lupaşcu and Butler (2019), in a more regional study in the United Kingdom, found results more aligned with our findings. Derwent and Parrish (2022) also showed results consistent with our findings, showing alignment between results derived from an observational-based analysis and modeled results (See Sect. S6 in the Supplement). "

E.4. Yet another under-discussed result is the influence of international shipping on European ozone. This is not mentioned in the Introduction section at all, and only one previous study is cited on this (Petetin et al., 2023), which is only cited in the conclusions section, with no discussion of why this study is significant. At the very least, there should be a paragraph in the Introduction on this subject, and the Results section should mention the previous literature where appropriate and discuss the new results in that context. There are also several other relevant studies here (Eyring et al., 2007; Karl et al., 2019; Lupaşcu and Butler, 2019; Butler et al., 2020; Jonson et al., 2020; Schwarzkopf et al., 2022; Zohdirad et al., 2022) as a non-exhaustive list.

As described in response E2, we added a new paragraph in the Introduction about the influence of international shipping on European O_3:

L76-86: "Shipping emissions also represent a notable source of O_3 in Europe. Lupaşcu and Butler (2019) estimated their contribution at approximately 19% to summer O_3 levels along the Mediterranean coast in 2010, while Pay et al. (2019) reported a 13% contribution during high O_3 episodes in 2012, particularly affecting the Mediterranean coastal areas of the Iberian Peninsula. Using a global model, Butler et al. (2020) showed the significant impact of NOx emissions from shipping on O_3 over the Northern Hemisphere oceans, contributing around 5-10 ppb along the Atlantic coast and 5-20 ppb along the Mediterranean coast on average in 2010. Jonson et al. (2020) and Schwarzkopf et al. (2022) showcased how shipping emissions contribute to O_3 titration near major shipping lanes in the North and Baltic Seas. Jonson et al. (2020) additionally reported that shipping contributes to higher O_3 levels along the Atlantic and Mediterranean coasts. Petetin et al. (2023) conducted a sensitivity analysis examining the impact of various emissions scenarios on O_3 pollution in Spain, finding that a 20% reduction in shipping emissions resulted in a 44% decrease in exceedances of the MDA8 O_3 threshold set by the AQGsv2021. This underscores the crucial role of maritime traffic emissions, particularly along the Mediterranean coast."

Furthermore, we have included additional discussions in the Results section highlighting the importance of international shipping:

L436-439: "This difference can be attributed to the favorable conditions for O_3 formation and accumulation in the Mediterranean, contrasting with the titration effect caused by elevated NOx concentrations at Europe's Atlantic principal ports (Amsterdam, Hamburg, Rotterdam, and London), as Jonson et al. (2020) and Schwarzkopf et al. (2022) pointed out."

L439: "In the latter, the SEA O_3 concentrations remain below 14 $\mu g/m^3$ (Fig. 6b). Lupaşcu and Butler (2019) found similar results, emphasizing the significance of shipping activities along the Mediterranean coast."

Detailed comments

E.5. Section 2.1: Are the ozone concentrations reported at the lowest model level, or are they extended to the surface? For maximum policy-relevance, ozone values at 2m should be reported. More information on the model vertical structure is needed here, including the height of the lowest model level.

The O_3 concentrations reported in our study are at the mid layer of the lowest model sigma level, which is approximately 20 meters above ground level. Although some numerical models compute a diagnostic to extend the lowest model level concentration to a specific height above ground level, CMAQ does not include this feature. We acknowledge this as a limitation but of second order compared with other uncertainties related with emission, transport, or chemistry processes in chemical transport models with an already shallow lowest model layer. The system used in this study was configured with 38 sigma layers extending from the ground up to 50 hPa in altitude, with 11 layers describing the PBL. We have clarified this in the revised manuscript as follows:

L127-130: "In this study, CALIOPE was configured to cover Europe at a horizontal resolution of 18 x 18 km2 with 38 sigma layers from the ground up to 50 hPa in altitude, where the mid-layer height of the lowest model level is set to 20 m above ground level. Approximately 11 layers characterize the planetary boundary layer (PBL)."

E.6. Lines 148-151: It would be useful to give some examples of the different kinds of tagging systems listed here. Also, there is one more option that is not listed here: simultaneous full attribution to both NOx and VOC reactants. This is the approach taken by Lupaşcu and Butler (2019), and it avoids the disadvantages of the pure forms of options 2 or 3, at the cost of increased difficulty of interpreting the results.

We appreciate the suggestion to provide examples of different O_3 tagging systems. Indeed, full attribution to both NOx and VOCs reactants was already listed as option 4. We rewrote this part to be more clear and with specific examples of the different tagging methods as follows:

L148-163: "O_3 source apportionment tagging methods employ a tracer species system to track O_3 and its precursors for specific emission categories (such as emission sectors) and/or geographical regions. Different source apportionment schemes have been proposed depending on the attribution perspective. These options include attribution based on

the reactant stoichiometry of all the species (1) (Shu et al., 2022), full attribution to NOx reactants (2) (Emmons et al., 2012; Butler et al., 2018; Lupaşcu and Butler, 2019; Butler et al., 2020; Shu et al., 2022; Romero-Alvarez et al., 2022), full attribution to VOCs reactants (3) (Butler et al., 2011, 2018, 2020; Lupaşcu et al., 2022; Shu et al., 2022), attribution to NOx and VOCs species (4) (Grewe et al., 2010, 2017; Shu et al., 2022), and attribution to NOx or VOCs reactants based on the ratio of chemical production of H_2O_2/HNO_3, which characterizes the chemical regime (5) (Valverde et al., 2016; Karamchandani et al., 2017; Pay et al., 2019). Each of these approaches has its unique characteristics and limitations, with different implications for air quality management and outcomes (Shu et al., 2022). For example, option 2 may result in an overestimation of NOx sources, as it attributes O_3 solely to NOx reactants. Conversely, option 3 may predominantly assign O_3 to biogenic emissions due to the significant amount of biogenic VOCs emitted compared to other sectors. Additionally, methods 1, 4, and 5 may have larger hemispheric contributions compared to methods 2 and 3, mainly due to the loss of label of NOx precursors as more reactions occur. In these cases, the high amount of hemispheric sources of O_3 or precursors could lead to a higher attribution to this source, as can be seen in Shu et al. (2022). In this work, we used an approach based on the limiting precursor of O_3 (5). Here, O_3 production is attributed to the limiting precursor responsible for O_3 formation."

E.7. Line 155: What are the implications of the limiting-precursor approach as chosen here in comparison to the other possible tagging method?

The chosen limiting-precursor approach explains the responsible of the O_3 formation, determined by the $PH_2O_2/PHNO_3$ ratio (ratio between the production rates of hydrogen peroxide and nitric acid). This approach is typically used in regional modelling studies, in which the resolution is higher compared to global models. Using this approach, instead of the others, helps us to understand better the role of each precursor (NOx and VOCs). In most of the cells and time, we will be in a NOx limited regime, where the O_3 production will be totally attributed to NOx precursors (similar to full attribution to NOx precursors method (2)), giving information on the role of NOx species (anthropogenic and biogenic). On the other hand, when we are in a VOCs limited regime, mainly close to areas with high NOx emissions, such as cities, O_3 production is attributed to VOCs precursors (similar to full attribution to VOCs precursors method (3)), which are essential in these areas to form

O_3. In this regime, we will find the main difference between method 2 and this method, where method 2 will attribute more O_3 solely to NOx precursors. Compared to method 1, 3 and 4, generally due to the high amount of biogenic VOCs precursors, they will have an attribution where the biogenic sources will dominate the O_3 production.

Additionally, methods 1, 4, and 5 have larger hemispheric contributions compared to methods 2 and 3, mainly due to the loss of label of NOx precursors as more reactions occur. In these cases, the high amount of hemispheric sources of O_3 or precursors could lead to a higher attribution to this source, as can be seen in Shu et al. (2022).

This discussion on the methods and their limitations has been introduced as described in the previous response in L156-163.

E.8. The method would be sensitive to the chemical regime as simulated by the model. How well does the model reproduce NOx-limited or VOC-limited regimes? How does it compare to other models in simulating the chemical regime?

Regarding the sensitivity of the method to the chemical regime biases simulated by the model, it's indeed an important aspect to consider. Our approach relies on either NOx or VOCs, meaning that uneven underestimation of these precursors could lead to leaning toward either NOx-limited or VOC-limited conditions. For example, Li et al. (2022) showed results with a greater underestimation of VOCs compared to NOx, potentially placing the chemical regime towards what's referred as transitional (when the O_3 production is shifted from one regime to the other), leading to a misattribution of the regime. Evaluating VOCs and NOx in Europe is challenging due to the limited monitoring stations of VOCs, making it complex to draw conclusions and assess model biases. However, we might expect a larger underestimation of VOCs than NOx similar to Li et al. (2022), implying our model regime shift would lean towards a VOCs-limited regime.

In our study, we use the CMAQ-ISAM method based on the limiting-precursor approach with a $PH_2O_2/PHNO_3$ threshold of 0.35 to identify a NOx or VOCs limited regime. The $PH_2O_2/PHNO_3$ value appears to be in the lower range compared to recent findings (Li et al., 2022), which would imply our attribution be shifted towards VOC limited regimes, but this ratio can vary significantly based on emissions and regional conditions. While other studies attempt to calculate different ratios as proxies for the chemical regime, such as $HCHO/NO_2$ or HCHO/NOy, which can be compared with satellite observations, uncertainty in this

calculation remains high.
Nevertheless, the impact of the potential misattribution of the regime in our study is more limited as O_3 production is attributed by country. Fig. 1 illustrates how the country-to-country contribution of NO_2 mainly affects border countries, pointing out that the majority of imported O_3 is transported rather formed from imported NO_2. This reduces the importance of the possible chemical regime misattribution in our study.

We haven't compared the chemical regime with other studies specifically due to the difficulty and inherent uncertainties in approaching the chemical regime characterization. While our study does not include a specific comparison, we acknowledge the importance of such analysis in understanding better the robustness and limitations of our approach. In this regard, we have acknowledged this uncertainty in the revised manuscript as follows:

L167-173: "Depending on the regime, either the NOx or the VOC tracers associated to each of the tagged sources are proportionally attributed to the total O_3. The chemical regime that controls O_3 production is determined using the ratio $PH_2O_2/PHNO_3$ (ratio between the production rates of hydrogen peroxide and nitric acid), where a ratio below 0.35 designates a VOC-sensitive regime, while a ratio above 0.35 signifies a NOx-sensitive regime (Zhang et al., 2009). However, there may be uncertainties associated with the relative proportion of NOx and VOCs simulated by the model and the threshold value (Li et al., 2022). Nevertheless, the impact of the specific threshold value adopted in our study is somewhat limited since O_3 production is attributed by country."

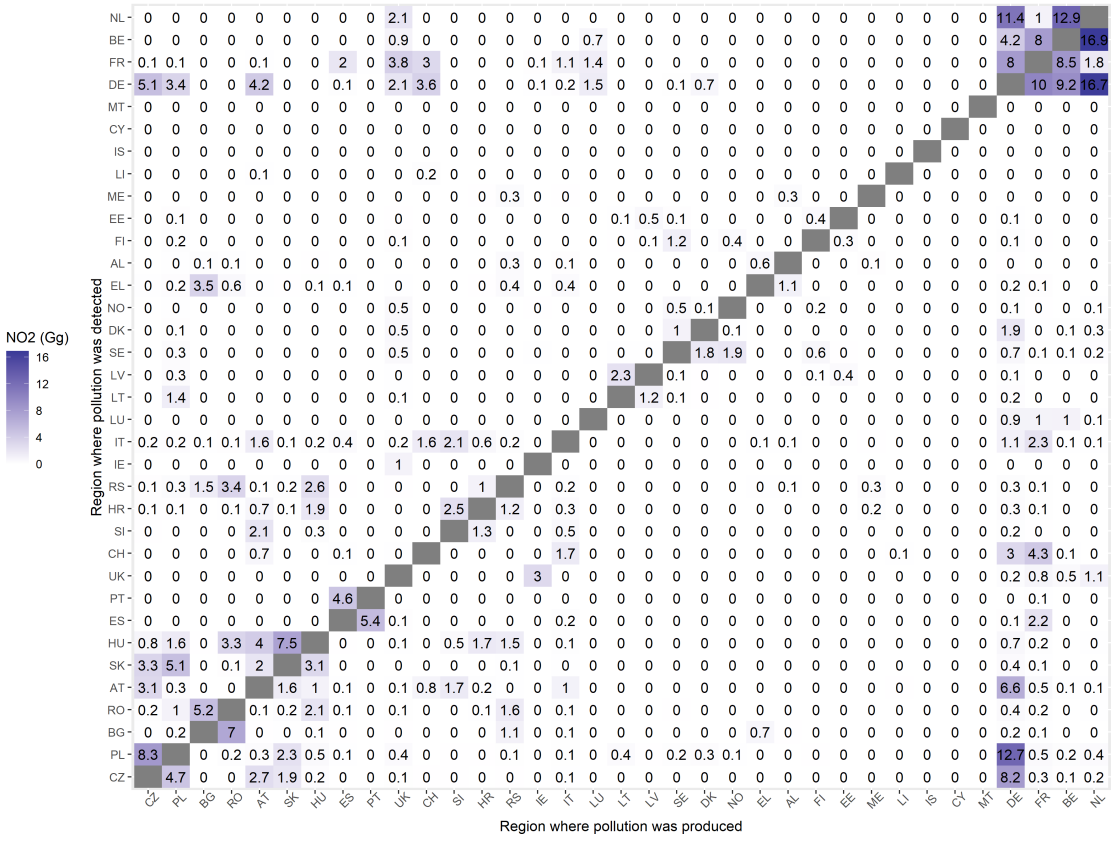

Figure 1: Country-to-country contributions to NO2 mass at each European country in summer 2015, 2016, and 2017.

E.9. In VOC-limited conditions, some fraction of the ozone production will be attributed to biogenic VOCs and methane (which has a longer lifetime but is ˜1000 times more concentrated than other VOCs).

> Certainly, in VOCs-limited conditions, the contribution of biogenic VOCs is expected to dominate other sources of VOCs, given the higher amount of BVOCs surrounding urban areas. It is important to emphasize that in our study, we have not differentiated the contribution by sector. Instead, we have analyzed them by country. However, it is within our future plans to disentangle the role of the biogenic VOCs and explore different tagging methods. New versions of CMAQ enable us to explore better this, and we aim to incorporate these aspects in forthcoming studies.

E.10. Line 164: Why does this method lead to a higher hemispheric contribution? A

short explanation would be useful here.

> Explanation in comment E7. We added a short sentence in the manuscript to clarify this point:
>
> L159-162: "Additionally, methods 1, 4, and 5 have larger hemispheric contributions compared to methods 2 and 3, mainly due to the loss of label of NOx precursors as more reactions occur. In these cases, the high amount of hemispheric sources of O_3 or precursors could lead to a higher attribution to this source, as can be seen in Shu et al. (2022)."

E.11. Line 165: How is methane handled in the ozone attribution? Is it contained in one of the species listed in Table S1? The CCAC Global Methane Assessment (CCAC, 2021) shows that ozone production is sensitive to methane in regions with high NOx, which includes much of Europe. How is ozone production from methane attributed in the current study?

> In our system, methane is not tagged, so O_3 production from methane is not attributed in this study. Table S1 in the Supplement lists the tagged species involved in O_3 formation within the CB05 extended mechanism. Methane concentrations are prescribed within CMAQ as a constant value of 1.85 ppmv in the mechanism and are not incorporated as emissions in the modelling framework. This method reflects a simplified approach to handle methane in the model. While we acknowledge the significance of methane, ongoing research is being conducted to understand its potential impact in regional models better.

E.12. Line 168: Does Figure 1 show the actual model domain? Latitude and Longitude scales should be added.

> Figure 1 is a representation of the different tags of the study. The model domain is shown in Fig. S1 with latitude and longitude. As suggested, we have added a clarification in Figure 1 caption as follows:
>
> L186 - Caption figure 1: "The model domain is shown in Fig. S1."

E.13. Line 172: Part of the ozone attributed to each country in this study will be from BVOC emissions, which may be beyond the control of policymakers. One might argue that this will only happen under VOC-limited conditions (where the excess NOx is from anthropogenic sources), but in this case, wouldn't it be better to attribute that ozone to the anthropogenic NOx emissions for the purposes of policy advice?

Certainly, it is important to acknowledge that a portion of the O_3 attributed to each country originates from BVOC emissions, which are beyond the control of policymakers. However, considering the overall balance of VOC (biogenic + anthropogenic) compared to NOx, it's likely that most of the time we are in NOx-limited conditions, and the O_3 will be attributed to NOx precursors. While our study does not specifically address the role of BVOCs, we plan to explore this aspect in forthcoming studies.

Attributing O_3 solely to anthropogenic NOx reactants emphasizes the role of anthropogenic NOx emissions, yet undervalues the role of both anthropogenic and biogenic VOC in the formation of O_3. On the other hand, attributing the O_3 formation to the limiting precursor answers the question, what drives O_3 formation?, which accounts for the importance of VOCs in NOx-saturated regime. All in all, different tagging approaches answer different questions, and can provide complementary information for policy advice. We also believe that more discussion in the modelling community is needed to better understand the implications of each method, and collaborative efforts can enhance the comprehensiveness of O_3 attribution studies.

E.14. Section 3.1: There is no discussion of how well the boundary condition from the global model represents the inflow of ozone into the model domain. Given that hemispheric transport is such a major contributor to ozone in this study, some discussion of this is needed, especially since there is a large inter-model spread in near-surface ozone from current state-of-the-art global models (Griffiths et al., 2021). Perhaps the authors could highlight the evaluation of their simulated ozone at the Mace Head station, which is primarily influenced by long-range transport.

We acknowledge the importance of evaluating background O_3 levels, especially in remote stations such as Mace Head, which is mainly influenced by O_3 long-range transport and has been extensively used to determine the background concentrations that arrive in Europe from the Atlantic Ocean (Derwent and Parrish, 2022). To address this comment, we analyzed the model performance in Mace Head, which provides further insight into the skills of the model to capture hemispheric long-range O_3 transport. Table 1 summarize the main statistics over the summers of 2015, 2016, and 2017 at Mace Head. The model exhibits a relatively low positive NMB and NME (below 15 %). All in all, the evaluation meets the metrics set by Emery et al. (2017) and Janssen et al. (2017), instilling confidence in the reliability of hemispheric background O_3 simulated by the model.

Table 1: Statistical overview of MDA8 O_3 concentrations over June-July-August 2015, 2016, and 2017 in Mace Head. The metrics are mean bias (MB), normalized mean bias (NMB), mean error (ME), normalized mean error (NME), and correlation coefficient (r). Also shown are the mean of the observations (MO), and the modelled mean (MM).

	MB (ppb)	NMB (%)	ME (ppb)	NME (%)	r	MO (ppb)	MM (ppb)
Mace Head	4.66	13.14	5.27	14.85	0.51	35.59	40.25

Some observation-based studies quantify the contribution of background ozone and regional photochemical production by analysing O_3 concentrations measured in stations representative of remote background conditions in contrast to stations also affected by local/regional photochemistry. This serves as an approximation to infer both the background and the photochemical enhancement over specific regions based on measurements. Although this method is not devoid of certain limitations, it can provide an estimate of the background O_3 contribution to total O_3. We have additionally expanded the evaluation of the model in the revised manuscript, introducing analyses disentangling background O_3 concentrations and photochemical enhancements.

We used O_3 concentrations obtained from remote and regional background EMEP stations across Europe. We specifically assessed the model's performance at stations where the simulated hemispheric contribution is at its peak, as well as stations where the model exhibits the greatest national influence (Fig. 2). This approach allowed scrutinizing the model's ability to capture background conditions and local/regional photochemical enhancements, respectively.

As depicted in Fig. 3, stations mainly influenced by background O_3 (Max BCON) are located along the Atlantic coast. On average there is a positive bias which remains below 17 %. For reference, our model closely aligns with the results obtained by the Copernicus Programme CAMS global reanalysis (CAMSRA), which is one of the best products available to describe O_3 concentrations at a global scale as it combines both model and observation-based information to provide a comprehensive estimation of the atmospheric conditions (Inness et al., 2019). On the other hand, the model presents a very good agreement in stations heavily influenced by local/regional photochemical production (Max National).

Most notably, this comparison highlights that overall the model very reasonably captures the transition from background conditions to photochemical O_3 production regimes, indicating its ability to reproduce O_3 variability effectively.

The good agreement between the background and photochemical increment derived from observations and the numerical simulations instills again stronger confidence in all the discussion and findings of our results.

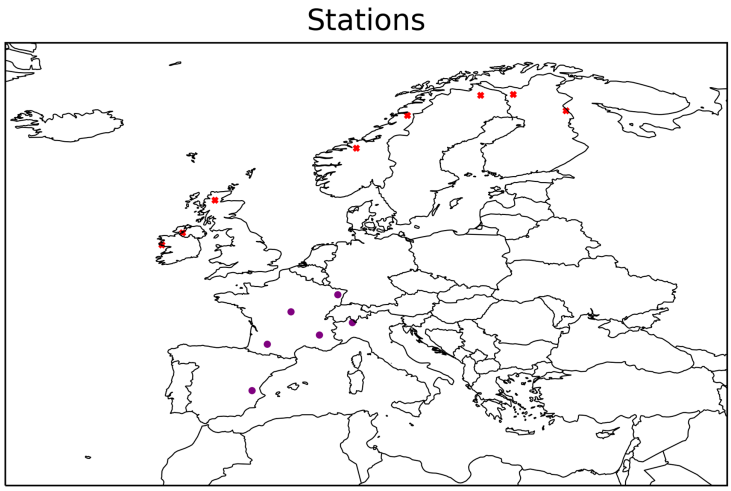

Figure 2: Geographical distribution of the stations with higher contributions from the hemispheric O_3 (red) and the stations with higher contributions from national photochemical O_3 production (purple).

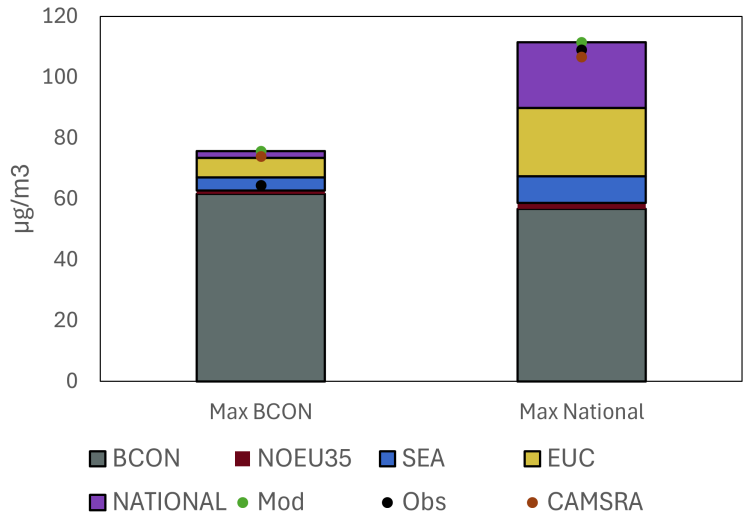

Figure 3: Average simulated source contributions to O_3 and total simulated and measured O_3 in EMEP stations where the contributions of hemispheric O_3 are above the 90^{th} percentile (Max BCON) and where the national contributions are above the 90^{th} percentile (Max National) of all EMEP stations. The geographical distribution of those stations is presented in Figure 2. Contributions include the national O_3 contribution (NATIONAL), the imported O_3 in each country originated from each of the remaining 34 countries (EUC), the contributions from hemispheric transport (BCON), non-European neighboring countries (NOEU35) and maritime sources (SEA). The green dot represents model total O_3 concentration, the black dot the O_3 concentrations from observations, and the red dot the O_3 concentrations from the CAMS reanalysis (CAMSRA).

We note that in addition to the previous analyses and in response to another reviewer, we have evaluated the regional photochemical contribution, and we compared our results against the observational-based analysis done by (Derwent and Parrish, 2022). Derwent and Parrish (2022) computed regional photochemical enhancement (RPE) values for the year 2020 at each of the low-elevation EMEP stations in northwestern Europe (32 stations). The RPE quantifies the photochemical production driven by local and regional anthropogenic precursor emissions. We conducted a comparative analysis of RPE values at these EMEP stations, derived from our simulations, with those reported by Derwent and Parrish (2022) (see Table 2). In our model-derived RPE estimation, we incorporate contributions from national sources, imported O_3 originating from the remaining 34 countries (EUC), and maritime sources (SEA). We note that while Derwent and Parrish (2022) computed RPE for the entire year 2020,

our study focuses exclusively on the summer months of 2015, 2016, and 2017. Therefore, consistent with the heightened RPE during summer conditions, we observe higher RPE values compared to those estimated by Derwent and Parrish (2022), whose RPE spans the entire year. Despite the disparity in analysis periods (annual vs summer), we find a robust correlation across the 32 stations between our simulated RPE and the observationally-derived values ($R^2 = 0.80$), as depicted in Fig. 4. Such comparison with observational data further substantiates the robustness of our modeling method to disentangle contributions from long-range transport and photochemical production driven by local and regional anthropogenic precursor emissions.

Two comprehensive sections have been included in the supplementary material, offering an in-depth analysis of both the background and regional photochemical contribution to the MDA8 O_3. See sections S5 and S6 in the revised manuscript supplement. Additionally, we rewrote the *Observations and model evaluation* and *Evaluation of simulated ozone concentrations* section to include the complementary evaluation described above:

L226-236: "Additionally, we specifically assessed the model's performance at stations where the simulated hemispheric contribution is at its peak, as well as at stations where the model exhibits the greatest national influence (Sect. S5). For this assessment we used the remote and regional background European Monitoring and Evaluation Programme (EMEP) stations (Tørseth et al., 2012) within the AQ (Air Quality) eReporting database of the EEA. This approach allows scrutinizing the model's ability to capture background conditions and local/regional photochemical enhancements, respectively. This comparison also uses as a reference the Copernicus Programme CAMS global reanalysis (CAMSRA), which is one of the best products available as it combines both model and observation-based information to provide a comprehensive estimation of the atmospheric conditions (Inness et al., 2019). Finally, we compared our results against the observation-based analysis of Derwent and Parrish (2022), where the contributions of long-range transport and regional photochemical enhancements (RPE) to O_3 were estimated for EMEP stations in northwestern Europe (see Sect. S6 for details)."

L263-268: "Additionally, the model captures well the increase in MDA8 O_3 concentrations between EMEP stations where the contributions from hemispheric transport O_3 is most dominant and those that are strongly impacted by national contributions, reflecting the ability of the model to represent local to regional photochemical O_3 enhancements (Sections S5 and S6 in the supplement).We also found a strong correlation between the observation-based estimates from Derwent and Parrish (2022) and our simulated values across 32 EMEP stations in northwestern Europe ($R^2 = 0.80$, see Sect. S6). Such comparison with observational data further substantiates the robustness of our modeling method to disentangle contributions from long-range transport and photochemical production driven by local and regional anthropogenic precursor emissions. "

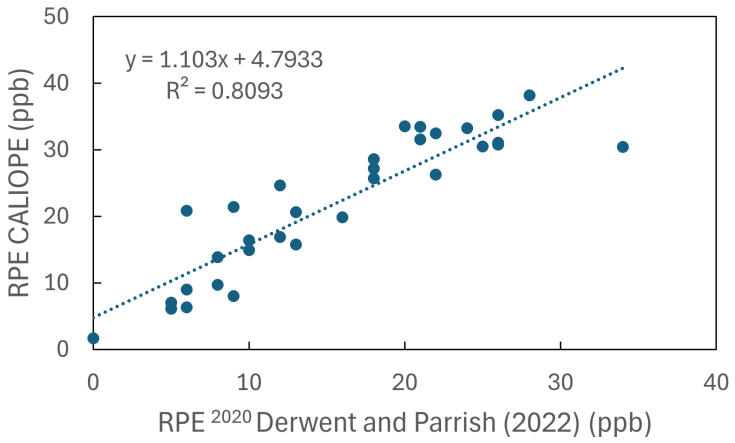

Figure 4: Correlation between the RPE found by Derwent and Parrish (2022) and by the CALIOPE system.

Table 2: Comparison of Regional Photochemical Enhancement (RPE) values from Derwent and Parrish (2022) and CALIOPE.

Station code	**Longitude,** °E	**Latitude,** °N	**RPE 2020** Derwent and Parrish (2022) (ppb)	**RPE CALIOPE** (ppb)
AT0002R	16.767	47.767	21	33.48
AT0042R	15.047	48.879	21	31.57
AT0043R	15.919	48.106	26	35.22
AT0045R	15.547	48.371	20	33.55
AT0046R	16.731	48.335	22	32.51
AT0047R	16.677	48.051	24	33.22
BE0032R	6.003	50.629	34	30.42
CH0002R	6.945	46.813	22	26.28
CH0003R	8.905	47.48	26	31.05
DE0001R	8.31	54.926	18	25.69
DE0002R	10.759	52.802	26	30.78
DE0007R	13.033	53.167	18	28.61
DE0009R	12.733	54.433	18	27.20
DK0031R	8.433	56.283	16	19.85
DK0041R	12.126	55.687	6	20.82
FI0009R	21.377	59.779	9	21.41
FI0017R	27.686	60.527	12	16.91
FI0022R	29.402	66.32	5	7.02
IE0031R	9.5	53.167	5	6.09
IT0004R	8.633	45.8	28	38.17
NL0009R	6.277	53.334	12	24.65
NL0010R	5.854	51.451	25	30.52
NO0001R	8.25	58.383	8	13.91
NO0015R	13.917	65.833	9	8.04
NO0039R	8.883	62.783	8	9.74
NO0042G	11.883	78.9	0	1.65
NO0043R	11.533	59	10	14.92
SE0011R	13.15	56.017	13	20.67
SE0012R	17.383	58.8	10	16.43
SE0013R	21.067	67.883	6	6.37
SE0032R	15.567	57.817	13	15.80
SE0035R	19.767	64.25	6	9.03

E.15. Line 244: Please check the grammar in this sentence, it doesn't seem to make sense to me.

We rewrote the text to clarify this sentence:

L272-274: "At the national scale, during the summers of 2015, 2016, and 2017, the countries' mean MDA8 O_3 concentrations range from 60-90 µg/m^3 in northern countries to 105-120 µg/mm^3 in southern countries (Fig. 2). "

E.16. Lines 390-400: This would be a good place in the manuscript to add some discussion of how these new results compare with previous work on the influence of international shipping on European ozone.

As described in response E4, we have extended the discussion of international shipping on O_3 concentrations based on our findings and available literature in the section suggested by the reviewer.

E.17. Line 413: The Gothenburg Protocol is not global. Its geographical scope is limited to the UNECE region.

Thank you for pointing this out. We changed "global scale" for "international scale" in the revised manuscript.

E.18. Lines 417-418: It is not clear from this text or from the caption to Figure 7 what exactly is shown here. Please explain this more clearly.

Figure 7 illustrates the NATIONAL contributions of the countries that exhibit the largest influence on the MDA8 O_3 levels across Europe. Each country's contribution depicted in the figure encompasses its impact on the overall MDA8 O_3 levels throughout Europe, including its own contribution. We rewrote the text as follows:

L462-464: "Fig. 7 shows the percentage contribution of the main contributors to European O_3 levels, including Germany, France, Italy, United Kingdom, Poland, and Spain (Fig. 3). Each country's contribution depicted in the figure encompasses its impact on the overall MDA8 O_3 levels throughout Europe, including its own contribution."

and Figure 7 caption as follows:

L494 - Caption Figure 7: "Figure 7. Contribution in percentage of the main countries with the highest MDA8 O_3 levels in Europe, average of summer 2015, 2016, and 2017. Each country's contribution depicted in the figure encompasses its impact on the overall MDA8 O_3 levels throughout Europe, including its own contribution. "

E.19. Line 447: The "potential to address" high ozone levels is problematic here. The authors seem to mean that if the contribution is high, then there must be a high potential for reduction, but this is not how policymaking works. Other factors are also important, such as whether the current emissions are due to the best available technology or not, what the costs of mitigation would be relative to the benefits of meeting regulatory targets, and whether or not any reductions would actually contribute to meeting these targets. I don't expect the authors to go into this level of detail, but I think they should define what they mean by "potential" here.

We agree that our statement may be ambiguous and not targeting all policy considerations. We rewrote the text to clarify this point:

L495-499: "In this section, we aim to identify both the main regions in Europe where there may be more potential to address high O_3 levels through national abatement plans and those that are more dependent on abatement plans of other European countries. Note that such potential could be limited by other factors not addressed in this section, such as whether the best available technologies are already in place, the cost of deploying specific mitigation actions, or whether the achieved reduction would actually contribute to meeting objective targets."

E.20. Line 473: Part of the reason why Western European countries tend to have a higher contribution to their own ozone levels could be due to the simple fact that they often tend to be larger, so that ozone production downwind of emission sources is more likely to take place within the national boundaries. This is not really discussed here.

We added a sentence acknowledging the limitation raised by the reviewer at the beginning of the paragraph to clarify this point.

L524: " An important aspect is the country size, where larger countries will tend to have larger ratios since O_3 production is more likely to take place within the national boundaries. "

E.21. Figure 9: The transition from ozone-exporting to ozone-importing countries appears to generally follow a west-east gradient. Countries in Western Europe still do import ozone, but because of their geographical location, this ozone is tagged as "BCON". The prevailing westerly winds then lead to ozone export from Western Europe

to Eastern Europe. This pattern can also be seen in Figure 2. The way that the authors present this ozone import/export is still useful as a kind of "blame matrix", but the role of geography on this result should be better discussed.

In this figure, we do not take into account either the BCON, the NOEU35, and the SEA, to provide an overall synthesized view of the contribution of each country and to better understand the role of each country to the O_3 in Europe. In this balance, only the European contribution is taken into account. We added a sentence to clarify this point.

L540-542: "This section analyses the balance between the imported and exported cumulative total mass (Tg) of O_3 at the surface among European countries, independent from individual NATIONAL contributions and contributions from the BCON, NOEU35 and SEA. "

Furthermore, the role of geography and how it affects the European O_3 levels are discussed along the manuscript and also in this section.

Ex: L553-558: "The major net importers of O_3 across Europe are Greece and Italy (net import balance above 0.5 Tg), each with very distinctive situations. Greece mainly imports O_3 (marginal export contribution) primarily due to its geographical location surrounded by high-emitter countries in the north and concentrated national emissions in the south of the country. On the contrary, Italy is somewhat protected by the orography in the north (as discussed in Sect. 3.5), imports large amounts of O_3 mainly from France, Germany, Spain, and the United Kingdom (Fig. **??**, while still exports large amounts of O_3 towards the Mediterranean neighbours."

Ex: L562-564: "The geographical localization of the United Kingdom (mainly affected by Atlantic Ocean air masses) and being one of the largest O_3 precursor emitters in Europe (see Table S7) explain this unique result."

E.22. Line 568: I do not understand this sentence. Please clarify this.

Once we normalize by the surface of each country, we saw that the main emitting countries (e.g. Germany, France, Spain,...) had the same range (import/export) as most European countries, indicating that these countries by the surface that they have, they have little impacts to the other European countries. So, the size of the countries compensates the O_3 mass export/import of the main emitting coun-

tries. We rewrote the text as follows:

L619-621: "Generally, there is a compensation of the O_3 mass with the surface area of the main emitting countries of precursors of O_3. This is primarily due to their larger surface area, leading to comparatively little impact on their neighboring countries."

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Response to Reviewer 1

We thank the reviewer for the insightful comment, which has helped us complement and improve the manuscript. A detailed response to all the questions, comments, and suggestions raised by the reviewer is provided below.

E.1. The latter comparison is quite persuasive to me, given that the results are derived from such different approaches. However, I must argue a bit with one paragraph in their rebuttal (which is also included in the revised Supplement).

"We note that while Derwent and Parrish (2022) computed RPE for the entire year 2020, our study focuses exclusively on the summer months of 2015, 2016, and 2017. Therefore, consistent with the heightened RPE during summer conditions due to higher temperatures and solar radiation, we obtain higher RPE values compared to those estimated by Derwent and Parrish (2022), whose RPE spans the entire year. However, despite the disparity in analysis periods (annual vs summer), we find a strong correlation across the 32 stations between our simulated RPE and the observationally-derived values (R2 = 0.80), as depicted in Fig. 3."

Derwent and Parrish do consider observational data from the entire year, but the RPE they calculate is based upon annual maximum 8-hourly (AM8) mean ozone mixing ratios. These annual maxima predominately occur in summer, so the differences that are seen in the comparison cannot really be attributed to the disparity in analysis periods (annual vs summer). I suggest that this paragraph in the Supplement be revised accordingly.

We appreciate the reviewer's attention to the relevant methodological aspects of Derwent and Parrish (2022) study, which are important for comparing RPE values derived from different analysis periods and metrics. In response, we have carefully revised the Supplement (Note 6 in the revised manuscript) clarifying the methodology followed by Derwent and Parrish (2022) and providing a more consistent explanation of our comparison. The text of the supplement now reads as follows:

L65-80: "Derwent and Parrish (2022) investigated long-term changes in the annual maximum 8-hourly (AM8) O_3 mixing ratios from 1989 to 2018 at 32 low-elevation EMEP stations in northwestern Europe. At Mace Head, two contributions were estimated by fitting their sum to the AM8 time series: a background O_3 transported into the region (TB), modeled with a three-term power series, and a regional photochemical enhancement (RPE), modeled as the 2000 RPE multiplied by an exponential decay function of time. The fitted TB contribution at Mace Head was then subtracted from the AM8 at each location, and the resulting RPEs were fitted

to the exponential decay function, representing the contribution of local and regional anthropogenic precursor emissions to O_3.

We compared the RPE fitted values from Derwent and Parrish (2022) with our simulations at these EMEP stations (Table S7). Due to the short duration of our study, we could not derive AM8 fitted values. Instead, our model-derived RPEs are based on the mean MDA8 for June-August 2015-2016, considering national sources (NATIONAL), imported O_3 from 34 other countries (EUC), and maritime sources (SEA). Our simulated RPEs show a strong correlation with the observationally-derived values from Derwent and Parrish (2022) across the 32 stations ($R^2 = 0.85$) (Fig. S8 a).

Despite using a different metric, the strong correlation between the AM8 fitted values from Derwent and Parrish (2022) and the mean MDA8 observed at each station (Fig. S8 b) demonstrates the relevance of our comparison. This comparison with observational analysis underscores the robustness of our modeling method in disentangling contributions from long-range transport and photochemical production driven by local and regional anthropogenic precursor emissions."

To support the revised section, we have included a new plot (Fig. S8 b) illustrating the strong correlation between the AM8 fitted values from Derwent and Parrish (2022) and the mean MDA8 observed at each station, which provides confidence with our comparison using different metrics (i.e AM8 vs MDA8). Finally, Table S7 has been updated, now presenting average RPE values for 2015, 2016 and 2017 consistently for both Derwent and Parrish (2022) and our model.

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Response to Reviewer 2

We thank the reviewer for the insightful comment. A detailed response to all the questions, comments, and suggestions raised by the reviewer is provided below.

E.1. The authors have done a good job of responding to the comments of both reviewers, and of revising their manuscript accordingly. The resulting manuscript is of high quality. I somewhat agree with the criticism of Reviewer 1, that the submission may not fully satisfy the stated scope of Communications Earth & Environment, that "Research papers published by the journal represent important advances bringing new insight to a specialized area of research." (quote from the Guide to authors on the journal website). None of the individual insights from the work represent especially important advances. Nevertheless, I also see value in the authors' response that the sheer breadth of their new work goes well beyond any of the individual studies that have been published on this topic. I would definitely like to see this work published, but I am not completely sure whether Communications Earth & Environment is the most appropriate journal for this. I would leave the final decision on this to the professional editorial team of the journal.

> We believe that our paper offers valuable insights that align well with the aims of *Communication Earth & Environment*. We would like to highlight the added value compared with other studies. Our paper stands out in the literature due to its comprehensive spatial and temporal coverage and detailed analysis of the transboundary O_3 contributions across 35 countries in Europe, encompassing aspects such as import-export dynamics, interannual variability, and the contributions of strong episodes, many of which have been barely explored in such depth in previous studies. We believe these contributions make our paper particularly compelling for the readers of *Communications Earth & Environment*.